

HEATING OF COAL WITH LIGHT PULSES

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Introduction

The carbonization of coal is normally carried out under conditions of relatively slow heating, the process taking many hours. During the past decade, interest has arisen in faster heating rates. Thus, the use of fluidized beds¹, circulating heated pebbles², and hot-gas³ carbonization treatment has reduced the heating periods into the time range of seconds or less. Investigators have reported in detail carbonizations in time ranges as low as 60 milliseconds³. Nelson reported the use of flash heating on coal samples but did not detail his findings⁴. His technique resulted in one to three milliseconds exposure. The present work was undertaken to obtain further information on the effect of flash-heating on coal using even shorter exposures.

Experimental

The flash unit consisted of a flashtube holder, four 100 μ F capacitors, and a 4000V power supply. A GE FT524-Xenon-filled quartz helix flashtube was used. The flash unit was incorporated into a gas-handling system as shown in Figure 1.

The duration of flash is a function of the electrical circuit, particularly the capacitance and the resistance of the flashtube. By definition, the flash duration is considered to be the time from the initial 1/3-peak power to the final 1/3-peak. With a given tube, the length of the flash varies with the capacitance. The flash duration was determined from data supplied by the manufacturer⁵, Table I.

TABLE I
Flash Characteristics

Capacity	Time above 1/3 power, μ sec.		Input, joule	Output, j/sq. cm. ^{4/}
	Max.	Min.		
100 μ F	230	195	800	3.7
200	-	305	1600	7.5
300	-	415	2400	11.2
400	800	520	3200	15

*The internal surface of the helix is 62 sq. cm.

The input energy dissipated in the flash was computed from the formula $E = 1/2 CV^2$, where E is the energy in joules, C is capacitance in microfarads, and V is voltage in kilovolts. Thus, the FT524 tube emits 15 optical joules/sq. cm. for an energy input of 3200 joules. But, as seen above, the light energy output is considerably less than the input energy. Approximately 30 percent of the input energy was radiated into the test cylinder.

The experimental system used is shown in Figure 1. Usually 10 mg. of pulverized coal (less than 10 microns in size) were transferred into the quartz reactor and suspended on the walls by rotating the reactor. The reactor was then carefully placed into the helix of the flashtube, connected to the manifold, and evacuated. After flashing at the desired energy level, the pressure rise was measured and a gas sample was taken. The residual solids were sampled or weighed when necessary.

As the characteristics of both the lamp and the quartz reactor tended to change with time, test series designed to achieve specific goals were run closely together.

Results

A series of coals having increasing volatile matter content were studied. Their compositions and proximate analysis are shown in Table II.

TABLE II
Composition of Coals Used

Coal	<u>Elkol¹</u>	<u>Federal No. 1²</u>	<u>Kopperston No. 2²</u>	<u>Colver²</u>
<u>Proximate Analysis,</u>				
<u>%, dry basis</u>				
Volatile Matter	40.7	37.7	31.6	25.3
Fixed Carbon	54.6	56.8	63.9	68.5
Ash (dry)	4.7	5.5	4.5	6.2
<u>Ultimate Analysis,</u>				
<u>%, dry basis</u>				
Carbon	70.6	78.4	85.1	80.6
Hydrogen	5.4	4.9	6.2	5.3
Sulfur	1.0	1.9	0.7	0.8
Nitrogen	1.2	1.5	1.5	1.5
Oxygen	17.1	7.8	2.0	5.6
Ash	4.7	5.5	4.5	6.2

¹Kemmerer Coal Co., Frontier, Wyoming

²Eastern Gas and Fuel Associates, Pittsburgh, Pa.

The composition of the evolved gases from the last three coals is shown in Figure 2 through 4. As the energy of the flash is increased, the gas composition changes. Hydrogen increases, the volume percentage of C_2H_2 remains nearly constant, while the more saturated hydrocarbons decrease. CO_2 decreases, while CO increases.

The gas-composition trends are consistent with increased cracking of the evolved gases with the higher temperatures associated with the increased energy. Thus, the more saturated ethane and ethylene are replaced by acetylene. Also, methane may crack to acetylene. However, the acetylene may decompose to carbon and hydrogen.

It is of interest to relate the product concentration to the volatile matter of the coal. The younger coals show less hydrogen and methane in the product gas. However, increasing the energy input increases the concentration of both of these gases, Figure 5.

Tests were also run to determine the effect of various atmospheres on the product distribution. As stated previously, most of the experiments were run in vacuum. In a departure from this usual practice, nitrogen was added to serve as an absorber for the high energy believed existent in the products. It is seen in Table III that 10 mm. of N_2 had no significant effect, when compared with the vacuum runs.

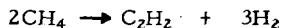
TABLE III

Effect of Increased Pressure

200- x 325-mesh Elkol particles, 3200 joules input

Gas Composition, mol %	10 mm N_2	Control (vacuum 0.1 mm)
H_2	44.5	42.4
CH_4	0.9	0.7
C_2H_2	20	20.7
CO	24.5	25

The effect of hydrogen in repressing the decomposition reactions was investigated next. Two pressure levels were investigated. It is seen in Table IV that increasing the H_2 pressure decreases the C_2H_2 while increasing the CH_4 . This is consistent with inhibiting the reaction.



Some saturation of the C_2H_2 could also have occurred. Increasing the pressure increased the effect. Two sets of control runs are quoted because the tests were run at different times and the physical conditions of the reactor, light, etc. had changed.

TABLE IV

Effect of Hydrogen

Elkol 5-10 μ , 3200 joules energy input

Gas Composition	Mole % (on H_2 -free basis)			
	Higher pressure comparison		Lower pressure comparison	
	160 mm H_2	control (0.1 mm vacuum)	10 mm H_2	control (0.1 mm vacuum)
CH_4	4.8	0.8	0.4	0.4
C_2H_2	9.5	25.0	25	37.8
CO	85	65.5	71	49

The effect of steam on the reaction was studied by adding a drop of water to the coal. As shown in Table V results are similar to those obtained with H_2 .

TABLE V

Effect of Water

Elkol, 3200 joules energy input

Gas Composition. mol %	With H_2O	Without H_2O
H_2	48	59
CH_4	2.6	1.0
C_2H_2	8.2	14.7
CO	30.1	21.4
C_2 's	4.9	2.7

In the above results, all changes were attributed to variation of the energy input. However, the exposure time varied concurrently. With the available set-up, it was not practical to vary the electrical circuitry. Mechanical screening of the coal proved most practical as a means of varying the energy at constant exposure time. The technique consisted of using a double quartz reactor and interposing three layers of 18- x 14-mesh metal screen between to cut down the adsorbed energy without affecting the time, Table VI. It is seen that the screen, by reducing the impinging energy, reduces cracking reactions.

TABLE VI

Effect of Energy Variation on Yield

Double Quartz Walls

Elkol coal, average particle size 5-10 μ

Electrical Energy Input	With Screen	Without Screen	Without Screen
Gas Composition. mol %	3200j	3200j	800j
H_2	16	59	44
CH_4	5.5	1.0	3.0
C_2H_2	4.2	14.7	12.0
CO	53	21.5	18
CO_2	7.7	0.4	2.3
C_2 's	8.9	2.7	6.6
C_3 's	2.0	-	1
C_4H_2	0.2	0.7	0.8

Mass balances around the reactor show recoveries of 74 to 110 percent. The weight of gas was determined from the pressure rise. The solid residue was washed into a Celite filter-aid bed for weighing. In none of the tests was there any evidence of tar formation. As seen in Table VII, the amount of gas produced varied randomly, reflecting poor reproducibility in exposing all of the coal to the flash. Some of the coal invariably fell to the bottom of the reactor during handling. Also, some coal obviously was blown from the reactor wall during the volatile-matter release.

TABLE VII

Mass Balances

All runs at 3200 joules energy input

Coal	Wt. Charged, mg.	Recovery, mg.		% Recovered
		Wt. Solid	Wt. Gas	
Elkol	10.6	5.4	3.2	81
Elkol (N ₂ run)	10.2	5.5	6.2	115
Elkol (H ₂ run)	10.4	6.9	4.0	105
Elkol (H ₂ run)	11.3	8.6	3.8	108
Johnstown	9.7	8.5	1.7	105
Johnstown	10.7	6.7	1.8	80
Johnstown	10.1	8.5	2.5	110
Powhattan	10.1	6.3	2.9	92
Powhattan	10.0	7.6	2.8	104
Illinois No. 6	10.4	8.8	1.4	98
West Virginia	10.3	4.8	2.8	74
West Virginia	9.7	6.0	2.6	89

Since the distribution of solids and gas did not give a realistic estimate of the percentage of coal volatilized, various other techniques for obtaining this parameter were investigated, but none were successful.

Microscopic examination of the residues showed the presence of carbon-black particles. The presence of these particles could be interpreted as being the residue of almost completely vaporized coal particles or the final product of a hydrocarbon-cracking sequence. A variety of tests were therefore devised to clarify this point.

1) In order to preserve as much of the hydrocarbons as possible, an even faster quench was attempted. A special reactor vessel with a central cooling thimble was constructed by suspending a small 4-inch test tube within

a larger one. The thimble was filled with either a dry ice-acetone mixture or liquid nitrogen. At first, the cold thimble was placed above the top of the flash tube. Black sooty material was found all over the condenser indicating that gas-phase decomposition could have occurred. The thimble was lowered until it was finally completely within the flash zone. When liquid nitrogen was used, the thermal shock shattered the glass apparatus. However, the sooty appearance of the condensed particles persisted.

2) Attempts were made to distinguish microscopically between unreacted coal, char and acetylene black. The fineness of the starting material made direct decision among the possibilities impossible. Attempts to produce very uniformly sized coal were essentially unsuccessful. It was finally decided to attempt direct comparison by photographing specific areas of slides before and after flashing. The results are shown in Figure 6. The A designation indicates that the picture was taken before flashing, the B, after flashing. The grid was superimposed on the slides during printing and serves to locate points of interest. It can be seen that large clumps of coal were blown off the slide by the flash. For example, compare Figure 6A - B areas 7D, 2F. Some glass was blistered as in Figure 6B, areas 8D, 9F. Important observations can be made in Figure 6A - B, areas 8D, 8E - F. Here particles can be identified before and after exposure (note in particular the particle shaped like the state of New Jersey in the upper center of 8D). The particles are essentially similar in shape before and after the flash, thus indicating that complete decomposition of the coal has not occurred. Obviously these coals were not screened from the flash, as any particle which could be seen by the camera should also have been exposed to the flash. The conclusion therefore must be that only partial vaporization or decomposition of the coal particles occurred. However, the exact magnitude cannot be determined since the coal particles expand during the coking process.

Conclusions

Flash heating is a practical technique to produce short periods of high temperature in coal particles. The percentage of coal volatilized could not be determined, but microscopic examination suggests that total vaporization did not take place. No liquids were recovered, but the gases, whether produced directly or indirectly by cracking of precursor tars, contained many valuable constituents.

Acknowledgment

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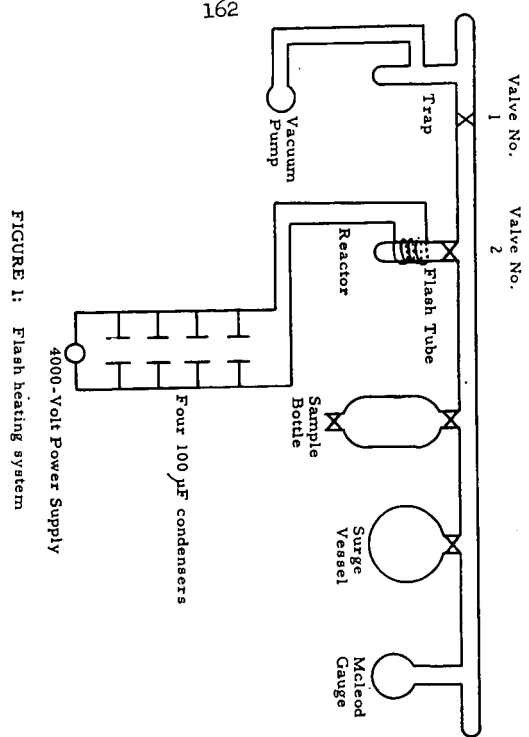


FIGURE 1: Flash heating system

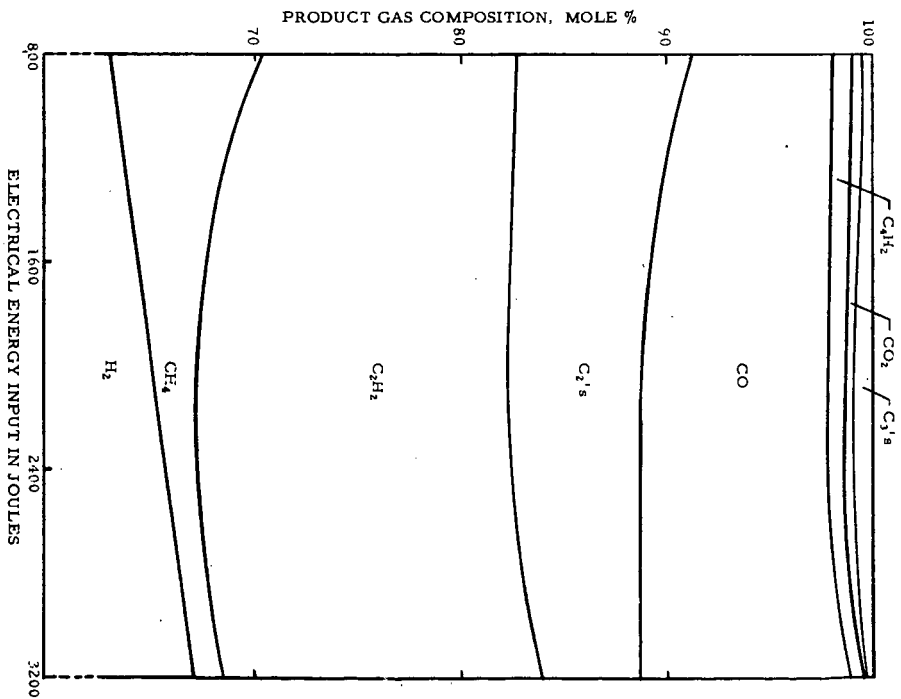


FIGURE 2: Gas composition from flash heating of Federal No. 1 coal (-400 mesh then pulverized), Quartz reactor

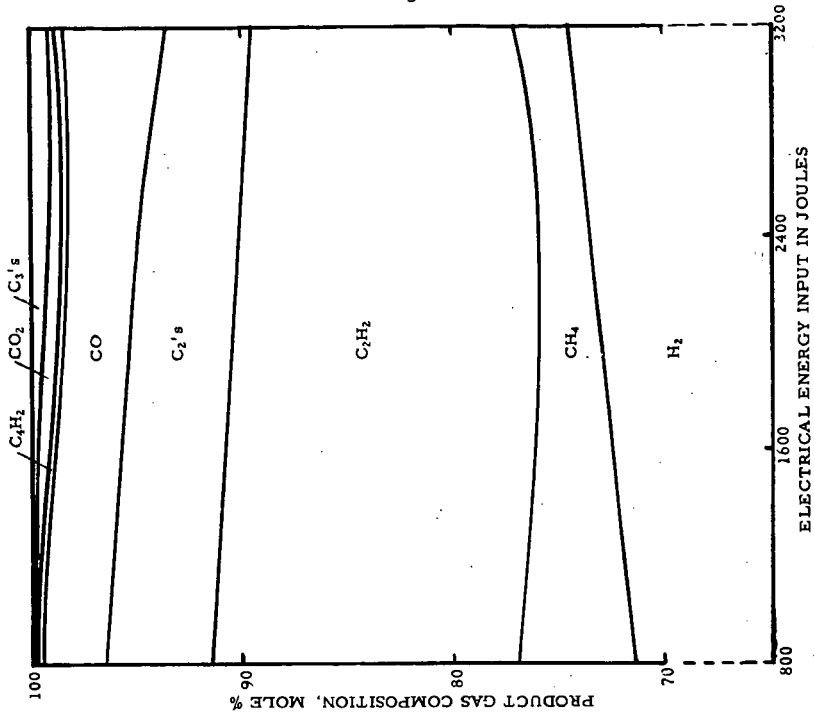


FIGURE 4: Gas composition from flash heating of Colver coal (-400 mesh then pulverized), Quartz reactor

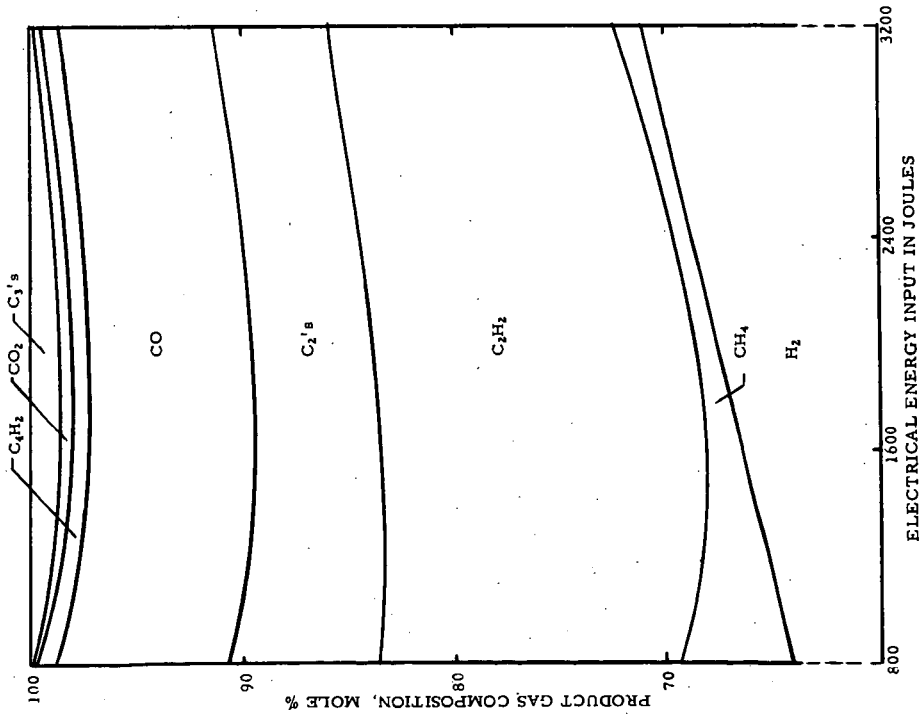


FIGURE 3: Gas composition from flash heating of Kopperston coal (-400 mesh then pulverized), Quartz reactor

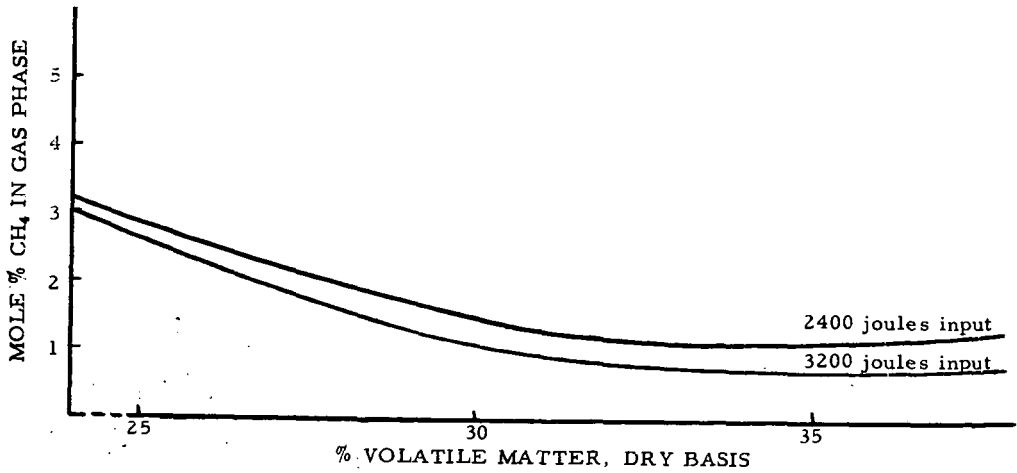
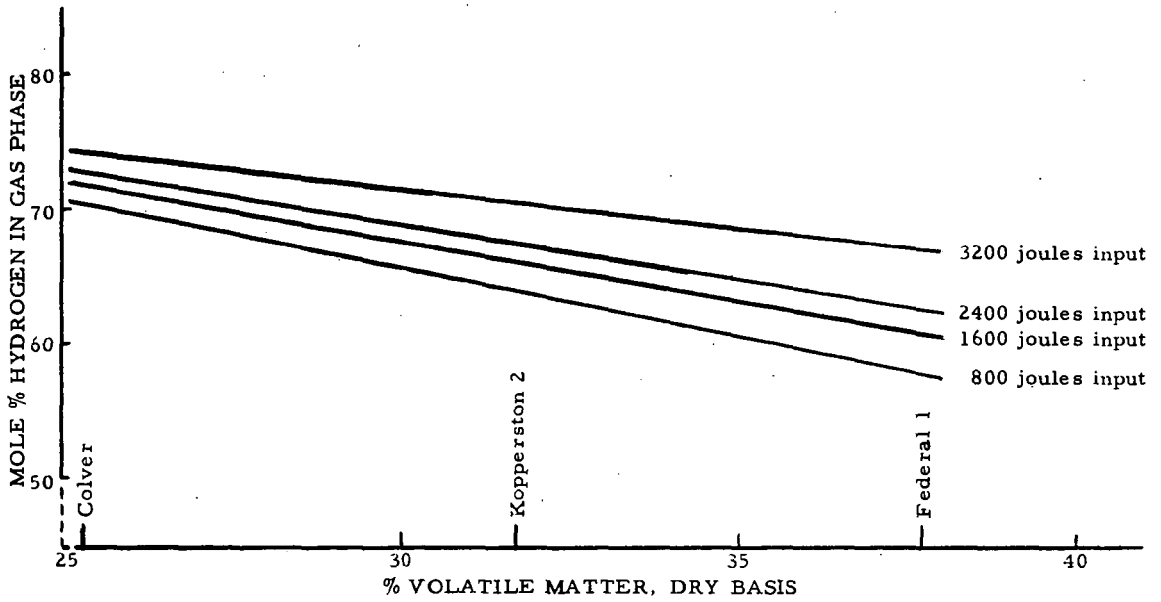


FIGURE 5: Relationship of H₂ and CH₄ concentrations to volatile matter of coals



FIGURE 6A: Before flash Each scale division = 2 microns
Energy of flash 3200 joules.
Elkol, powdered, then twice elutriated to separate out
the fines; average particle size about 5 microns.



FIGURE 6B: After flash Each scale division = 2 microns
Energy of flash 3200 joules.
Elkol, powdered, then twice elutriated to separate out
the fines; average particle size about 5 microns.